Preface

Statistical physics in soft matter systems, physical properties of bio-inspired systems and the mechanical manipulations of nano-systems have been studied using optical tweezers to form the basis of this doctoral Thesis. The first two chapters are on a general introduction about optical tweezers and detailed description of the setup used along with its calibrations. The next three chapters describe studies of statistical properties in soft matter systems, namely, out-of-equilibrium microrheology in a worm-like micellar system, irreversibility to reversibility crossover in the non-equilibrium trajectories of an optically trapped particle with the verification of fluctuation theorems even for non-ergodic descriptions of the system and high velocity Brownian vortexes at the liquid-air interface. The mechanical manipulation of the nano-systems, i.e. optically driven nano-rotors and the trapping, as well as transportation of palladium decorated single wall carbon nanotubes using optical tweezers have been discussed in the next two chapters. In the next chapter, the study of physical property of a bio-inspired system - the cell membrane deformability of human erythrocytes with increasing calcium ion concentration has been described. This Thesis is an endeavor to understand different mesoscopic systems using optical trapping and manipulation.

Chapter 1 gives an introduction on optical tweezers. The working principle of optical trapping and manipulation are discussed along with their applicability in different fields of physics.

Chapter 2 discusses the experimental setup in detail. The setup used for the experiments is a dual optical trap around an inverted microscope. The formation of the traps, the technique to steer the trapping beams and to place the traps at the desired positions in 3D without affecting the symmetry or stiffness are described. Instantaneous position tracking of the trapped particle is a very crucial part of optical trapping experiments. A tracking beam is used for this purpose and the trapped bead is imaged on a quadrant photo diode which provides the current signals that corresponds to the particle's position in the focal plane. Then the calibration of the setup using various calibration methods are explained. Calibration of the setup includes the calibration of the position sensing devices, e.g. the quadrant photo diode and the CCD camera attached to the microscope, calibration of the electronic devices, e.g. the stage nano-positioner, nano-tilt mirror mount etc., and finally calibration of the trap stiffnesses (in both X and Y) at varying laser powers. Precautions taken during the experiments to minimize the artifacts are also mentioned.

In **Chapter 3**, a nonlinear microrheology experiment to probe directional viscoelasticity of a sheared worm-like micellar system has been described. Many wormlike micellar systems exhibit appreciable shear thinning due to shear induced alignment. As the micelles get aligned, introducing directionality in the system, the viscoelastic properties no longer remain isotropic. An optical tweezers based technique enables us to probe the out-of-equilibrium rheological properties of CTAT (cetyltrimethylammonium tosylate, cationic surfactant) system simultaneously along two orthogonal directions - parallel to the applied shear, as well as perpendicular to it. A trapped bead is dragged through the medium (1 wt% CTAT) and the position fluctuations of the bead, along the direction of motion (X) and perpendicular to it (Y), are recorded in both 'drive on' and 'drive off' states. While the displacement of the bead along X - in response to the active drag force - carry signature of conventional shear thinning, its spontaneous position fluctuations along Y, following the fluctuation dissipation theorem, provide the loss modulus (G'' along Y) which manifests a dramatic orthogonal shear thickening, an effect hitherto unobserved.

Chapter 4 describes an irreversibility to reversibility crossover in the transient response of a particle in optical trap; and the verification of the fluctuation theorem for a non-ergodic description of this system. The transient position fluctuations of a colloidal bead is studied as it approaches equilibrium after being released from varying heights (by using an additional very strong optical trap) in the potential energy landscape created by a weak optical trap. The time evolution of the system shows dramatic changes as the release point energy is decreased. Starting from a small-time-reversible to long-time-irreversible transition for a higher energy release, a time independent completely reversible state could be reached just by lowering the initial potential energy a bit. For an even lower energy release, the system shows an anomalous irreversibility. In this state, it progressively extracts useful work from the thermal fluctuations and surprisingly goes to a higher energy phase point. Highlighting the competition between the micro-reversibility and the irreversible dissipative loss in determining the long-time system behavior, this study exhibits the prominent emergence of a completely reversible state even at long time, in between the two irreversible states of opposite kind. The Transient Fluctuation Theorem (TFT) and the Integrated Transient Fluctuation Theorem (ITFT) which are defined to be valid only for ergodic systems, have been verified even for non-ergodic descriptions (separately for different release points) of this system.

Chapter 5 illustrates the study of high velocity Brownian vortex at the liquid-air interface. A general kind of Brownian vortexes are constituted by applying an external non-conservative force field to a colloidal particle bound by a conservative optical trapping force at a liquid-air interface. As the liquid medium is translated at a constant velocity with the bead trapped at the interface, the drag force near the surface provide enough rotational component to bias the particle's thermal fluctuations in a circulatory motion. The frequency of that circular motion increases linearly with the stage velocity, while an increment in the trapping laser power shows the opposite effect. The properties of these Brownian vortexes have been studied extensively to demonstrate how the thermal fluctuations and the advection of the bead play their role in the vortex motions, with an inference that the angular velocity of the circulatory motions offer a comparative measure of the interface fluctuations.

In **Chapter 6** the optical manipulation of asymmetric nanorods that constitutes optically driven nanorotors are described. The light force, irrespective of its polarization, is used to run a simple nanorotor. While the gradient force of a single beam optical trap holds an asymmetric nanorod, the scattering force is utilized to generate a non-zero torque on the nanorod making it rotate about the optic axis. The inherent textural irregularities or morphological asymmetries of the nanorods give birth to chirality which is responsible for generation of the torque under the radiation pressure. A farther study on nanorotors that are more transparent to infra-red (trapping beam) confirms that the scattering force is indeed the origin of the torque. A model is proposed to explain the rotational motion of the nanorods and estimate the speed of rotation. If the nanorods are not fairly transparent to the laser beam, even a small surface irregularity with non-zero chirality is sufficient to produce enough torque for moderate rotational speed. Different sized rotors can be used to set the speed of rotation over a wide range, with fine tuning possible through the variation of the laser power.

Chapter 7 discuses optical trapping and transportation of palladium decorated single wall carbon nanotubes (Pd-SWNT). Individual carbon nanotubes being substantially smaller than the wavelength of light are not much responsive to optical manipulation. Decorating those single-walled carbon nanotubes with palladium particles changes that scenario dramatically, making the optical trapping and manipulation much easier. Palladium decorated nanotubes (Pd/SWNTs) have higher effective dielectric constant and are trapped at much lower laser power level with greater ease. In addition to that, an asymmetric line trap makes it possible to transport the Pd decorated SWNTs to a desired distant location in the sample cell. In the asymmetric line trap the Pd/SWNTs are first get attracted by the gradient force and then the scattering force push them away towards the other end of the line trap.

In **Chapter 8**, how the rotational motion of crenated erythrocytes in an optical trap can be used to probe their membrane deformability is explained. When placed in a hypertonic buffer medium, discocytic human erythrocytes are subjected to crenation and take deformed shapes. The deformation of the cells brings in chirality and asymmetries in shape that make them rotate under the scattering force of a linearly polarized optical trap. A change in the deformability of the erythrocytes, due to any internal or environmental factor, is reflected in the rotational speed of the trapped crenated cells. Therefore the average rotational speed and the probability of rotation of the crenated erythrocytes in an optical trap can be considered as a direct signature of their membrane deformability. As an example, the relative increment in erythrocyte membrane rigidity with adsorption of Ca^{++} ions is examined quantitatively through this approach.

The Thesis concludes with a summary of the main results and a brief discussion of the scope of future work in **Chapter 9**.